

Atmospheric Measurements and the Indirect Climatic Effects of Aerosols

John H. Seinfeld
California Institute of Technology
Pasadena, California 91125
(626) 395-4635 (626) 796-2591 seinfeld@caltech.edu

Award Number: N00014-96-1-0119

LONG-TERM GOALS

The long-term goal of this project is to gain a deep understanding of the role of atmospheric aerosols in affecting transmission of radiation through the atmosphere and in altering global climate.

OBJECTIVES

The scientific objectives of this project are to identify the specific manner in which atmospheric aerosols affect the Earth's climate. The technological objectives are to develop state-of-the-art instruments for aircraft sampling of aerosols that advance the long-term goals of the project.

APPROACH

The main technical approach is to conduct aircraft studies of the atmosphere, in which comprehensive sampling of atmospheric particles and radiative and cloud properties is carried out. The aircraft studies are complemented by laboratory investigations and theoretical analysis. Key individuals participating in this work are Professors John H. Seinfeld and Richard C. Flagan at the California Institute of Technology and Dr. Haf Jonsson at Naval Postgraduate School. Professor Seinfeld serves as Principal Investigator. Professor Flagan plays a key role in instrumentation development and planning of aircraft operations. As Chief Scientist of CIRPAS, Dr. Jonsson oversees all aspects of aircraft measurements and data management.

WORK COMPLETED

During the past year, the work completed consists of the following:

1. Analysis of data from ACE-Asia field experiment (Japan)
2. Analysis of data from Cloud Halo field experiment (Oahu, HI)
3. Conducting Cloud Halo field experiment (Key West, FL)
4. Conducting Crystal-Face field experiment (Key West, FL)

Report Documentation Page			Form Approved OMB No. 0704-0188		
Public reporting burden for the collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to a penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.					
1. REPORT DATE 30 SEP 2002		2. REPORT TYPE		3. DATES COVERED 00-00-2002 to 00-00-2002	
4. TITLE AND SUBTITLE Atmospheric Measurements and the Indirect Climatic Effects of Aerosols			5a. CONTRACT NUMBER		
			5b. GRANT NUMBER		
			5c. PROGRAM ELEMENT NUMBER		
6. AUTHOR(S)			5d. PROJECT NUMBER		
			5e. TASK NUMBER		
			5f. WORK UNIT NUMBER		
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) California Institute of Technology,,Pasadena,,CA, 91125			8. PERFORMING ORGANIZATION REPORT NUMBER		
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)			10. SPONSOR/MONITOR'S ACRONYM(S)		
			11. SPONSOR/MONITOR'S REPORT NUMBER(S)		
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT The long-term goal of this project is to gain a deep understanding of the role of atmospheric aerosols in affecting transmission of radiation through the atmosphere and in altering global climate.					
15. SUBJECT TERMS					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT Same as Report (SAR)	18. NUMBER OF PAGES 9	19a. NAME OF RESPONSIBLE PERSON
a. REPORT unclassified	b. ABSTRACT unclassified	c. THIS PAGE unclassified			

RESULTS

Cloud Halo Field Experiment, Hawaii

Regions of enhanced humidity in the vicinity of cumulus clouds, so-called cloud halos, reflect features of cloud evolution, exert radiative effects, and may serve as a locus for new particle formation. We report here the results of an aircraft sampling campaign carried out near Oahu, Hawaii from July 31 through August 10, 2001 and aimed at characterizing the properties of trade wind cumulus cloud halos. An Aerodyne Research Inc. fast spectroscopic water vapor sensor, flown for the first time in this campaign, allowed characterization of humidity properties at 10 m spatial resolution. Statistical properties of 60 traverses through cloud halos over the campaign were in general agreement with measurements reported by Perry and Hobbs (1996). Cloud halos tend to be broad at lower levels and narrow at upper levels and broader on the downshear side than on the upshear side, resulting from turbulent mixing at the cloud boundary and cloud evolution. Aerosol measurements in the vicinity of the clouds and halos showed the typical bimodal distribution in the marine boundary layer and unimodal distribution in the free troposphere; newly formed particles, when present, were seen on the downshear side of the cloud. One particularly long-lived cloud is analyzed in detail, through both airborne measurement and numerical simulation to track evolution of the cloud halos over the cloud's lifetime. Radiative calculations carried out on the simulated cloud and halo field indicate that the average shortwave absorption as a result of the presence of halo regions is about 0.3% of total solar absorption in the column. The halo effect therefore represents only a small fraction of the 6% "anomalous cloud absorption" discrepancy identified in the atmospheric science literature.

Crystal-Face Field Experiment

During the Key West deployment in the summer of 2002, the payload on the CIRPAS Twin Otter included ideal instrumentation for an aerosol/CCN closure study (Table 1). Throughout the mission, the ambient aerosol was measured simultaneously by the Caltech CCN Counter, the DACADS instrument, which measures the aerosol size distribution, and the Aerodyne Aerosol Mass Spectrometer (AMS), which provides detailed chemical information for larger particles. The data from the AMS indicated that in most circumstances the aerosol was primarily ammonium sulfate; assuming this composition allows a straightforward comparison between the measured CCN concentration and a predicted concentration based on the aerosol size distribution. To make this comparison, the CCN concentration was averaged for the duration of a single scan of the dry aerosol size distribution from the DACADS instrument. This result was compared to the concentration of particles above a given cut size; this cut size varied slightly over the course of the mission, but could be accurately predicted using a model that simulated instrument performance using in-flight housekeeping data.

The results of the closure study are shown in Figure 1. Observed concentrations ranged from 70 to 6000 CCN/cm³ and there is very good agreement between the predicted and observed concentrations over the entire range. The agreement is consistent over most of the 17 data flights included in the figure, but there are occasions where there appears to be substantial undercounting, most notably on the July 28 flight. On the occasions of poorer agreement, there is evidence of strong anthropogenic influence, in the form of elevated particle counts, or elevated CO levels. The consistent agreement in most cases despite the assumption of ammonium sulfate is an indication that the discrepancies between measured and observed CCN in other studies may be due to measurement biases rather than insufficient understanding of activation processes.

A closure study between aerosol and warm-cloud microphysics was performed using field data collected during the NASA CRYSTAL-FACE campaign. CRYSTAL-FACE was conducted in continental and marine environments near southern Florida in July, 2002. Detailed profiles of thirteen cumulus clouds were made by the CIRPAS Twin Otter aircraft, which was equipped with four aerosol sizing systems, two CCN counters operated at 0.4% and 0.7% supersaturation, an Aerodyne aerosol mass spectrometer, a MOUDI filter sampler system, two cloud drop sizing probes, and two turbulence probes. A wide range of CCN (300 to $>3500\text{ cm}^{-3}$) and cloud drop concentrations (200 to $>1600\text{ cm}^{-3}$) provided an ideal case study for aerosol-cloud interactions and the first and second indirect effects. Vertical characterization of the young and mature cumulus clouds were obtained from multiple horizontal passes from below cloud base to cloud top. Excellent closure is obtained between a detailed adiabatic cloud activation model and the cloud drop concentration observed 100 m above cloud base. The model is constrained by observed updraft velocity and below-cloud aerosol properties (i.e. concentration, size distribution, composition, and supersaturation spectrum). Each cloud contained a core often exceeding 500 m in height in which the equivalent potential temperature follows a moist-adiabatic vertical profile. Effective radius most often follows an adiabatic profile, even in regions where liquid water content and/or equivalent potential temperature are sub-adiabatic. Large cloud-to-cloud variations in the vertical profile of effective radius are primarily driven by below-cloud aerosol concentration and to a lesser degree by cloud dynamics (i.e. vertical velocity). These data provide excellent constraints for studies using large-eddy-simulation models to understand the indirect effect of aerosols on cloud albedo, cloud lifetime, and precipitation formation in warm cumulus. Likewise, these Twin Otter data play key role in the larger CRYSTAL-FACE program objectives (which included of six aircraft and two surface stations) by providing insight into the influence of aerosol/warm cloud interactions on large-scale convective processes, such ice nucleation, mixed phase processes, cirrus albedo, and the thermodynamic forcing of large cumulus systems.

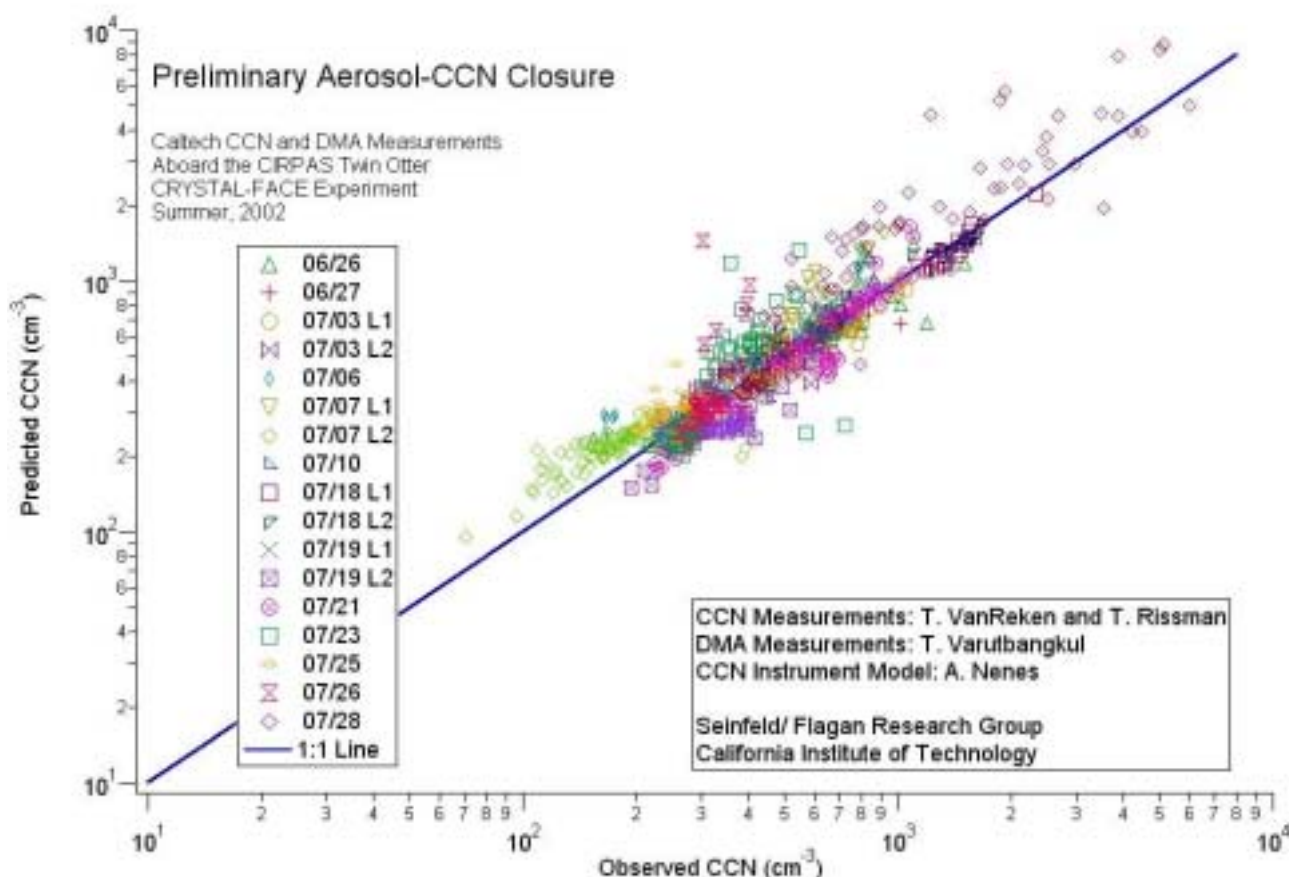


Figure 1. Comparison of measured CCN concentrations with predicted concentrations based on simultaneous data from a DMA. The effective supersaturation of the CCN counter was approximately 0.8%, and the aerosol composition was assumed to be pure ammonium sulfate. Each point represents one DMA scan compared against the average of CCN measurements taken during that scan. All data are from the Key West deployment of the CIRPAS Twin Otter. The legend gives the date for each flight; “L1” and “L2” indicate Leg 1 or 2 on a given day. Only data from level flight legs are included.

IMPACT/APPLICATIONS

The potential future impact of this project is to reduce the uncertainty in understanding the role of aerosols in the marine atmosphere and in estimating the future effects of anthropogenic and natural atmospheric aerosols on the Earth’s climate.

TRANSITIONS

A major future transition will be the use of hardware developed in this project by others. We anticipate such transitions may occur during the current grant period.

RELATED PROJECTS

A closely related project is the full Crystal-Face experiment, in which we participated. The appropriate web site is: <http://cloud1.arc.nasa.gov/crystalface>

SUMMARY

In the previous year we successfully completed two major field experiments. Work is ongoing on data analysis from both missions.

PUBLICATIONS

Collins, D. R., A. Nenes, R. C. Flagan, and J. H. Seinfeld, "The Scanning Flow DMA," *J. Aerosol Sci.*, **31**, 1129-1144 (2000).

Chuang, P. Y., A. Nenes, J. N. Smith, R. C. Flagan, and J. H. Seinfeld, "Design of a CCN Instrument for Airborne Measurement," *J. Atmospheric and Oceanic Technology*, **17**, 893-907 (2000).

Nenes, A., S. Ghan, H. Abdul-Razzak, P. Chuang, and J. H. Seinfeld, "Kinetic Limitations on Cloud Droplet Formation and Impact on Cloud Albedo," *Tellus*, **53**, 133-149 (2001).

Nenes, A., P. Y. Chuang, R. C. Flagan, and J. H. Seinfeld, "A Theoretical Analysis of Cloud Condensation Nucleus (CCN) Instruments," *J. Geophys. Res.*, **106**, 3449-3474 (2001).

Mader, B. T., R. C. Flagan, and J. H. Seinfeld, "Sampling Atmospheric Carbonaceous Aerosols using a Particle Trap Impactor/Denuder Sampler," *Environ. Sci. Technol.*, **35**, 4857-4867 (2001).

Collins, D. R., R. C. Flagan, and J. H. Seinfeld, "Improved Inversion of Scanning DMA Data," *Aerosol Sci. Tech.*, **36**, 1-9, (2002).

Roberts, G. C., A. Nenes, J. H. Seinfeld, and M. O. Andreae, "Impact of Biomass Burning on Cloud Properties in the Amazon Basin," *J. Geophys. Res.* (in press).

Lu, M.L., J. Wang, A. Freedman, H.H. Jonsson, R.C. Flagan, R.A. McClatchey, and J.H. Seinfeld, "Analysis of Humidity Halos Around Trade Wind Cumulus Clouds," *J. Atmos. Sci.*, (submitted for publication).

Wang, J., R.C. Flagan, J.H. Seinfeld, H.H. Jonsson, D.R. Collins, P.B. Russell, B. Schmid, J. Redemann, J.M. Livingston, S. Gao, D.A. Hegg, E.J. Welton, and D. Bates, "Clear-Column Radiative during ACE-Asia: Comparison of Multiwavelength Extinction Derived from Particle Size and Composition with Results from Sunphotometry," *J. Geophys. Res.*, (in press).

Nenes, A., R.J. Charlson, M.C. Facchini, M. Kulmala, A. Laaksonen, and J.H. Seinfeld, "Can Chemical Effects on Cloud Droplet Number Rival the First Indirect Effect?" *Geophys. Res. Letters*, (in press).

Mader, B. T., R. C. Flagan, and J. H. Seinfeld, "Airborne Measurements of Atmospheric Carbonaceous Aerosols During ACE-Asia," *J. Geophys. Res.*, (in press).

Conant, W. C., A. Nenes, and J. H. Seinfeld, "Black Carbon Radiative Heating Effects on Cloud Microphysics. and Implications for the Aerosol Indirect Forcing: 1. Extended Köhler Theory," *J. Geophys. Res.*, (in press).

Nenes, A., W. C. Conant, and J. H. Seinfeld, "Black Carbon Radiative Heating Effects on Cloud Microphysics and Implications for the Aerosol Indirect Effect: 2. Cloud Microphysics," *J. Geophys. Res.*, (in press).

Wang, J., R. C. Flagan, and J. H. Seinfeld, "A Differential Mobility Analyzer (DMA) System for Submicron Aerosol Measurements at Ambient Relative Humidity," *Aerosol Sci. Tech.*, (in press).

Wang, J., R. C. Flagan, and J. H. Seinfeld, "Diffusional Losses in Particle Sampling Systems Containing Bends and Elbows," *J. Aerosol Sci.*, **33**, 843-858 (2002).

Wang, J., V. F. McNeill, D. R. Collins, and R. C. Flagan, "Fast Mixing Condensation Nucleus Counter: Application to Rapid Scanning Differential Mobility Analyzer Measurements," *Aerosol Sci. Tech.*, **36**, 678-689 (2002).

PATENTS

None

Table 1. CRYSTAL-FACE Flight and Instrument Performance Summary

Last updated: 9/16/02

General Flight Information				Cabin Instruments										CIRPAS Instruments								
Flt #	Flight Date	Flt Time (UTC)	Flt Type (# of clouds/ reps)	CCN - CIT	CCN - SIO	DMA	AMS	CO	NOAA H ₂ O	MACS	SSFR	BSR	LEISA	ARI H ₂ O	MET	NAV (C,N,T)	CPC's (3,7,12)	APS	CAPS	FSSP	PCASP	MOUDI
Halo 1	6/24/02	~16:20-19:50	C(1)	down	N/A	OK	OK	N/A	N/A	N/A	N/A	N/A	N/A	OK	OK	so-so (C)	OK	OK	OK	OK	OK	N/A
Halo 2	6/25/02	~15:40-20:00	C(3), S	so-so	N/A	OK	OK	N/A	N/A	N/A	N/A	N/A	N/A	OK	OK	so-so (C)	so-so (3)	OK	OK	OK	OK	N/A
Halo 3	6/26/02	~16:05-20:00	C(1)	OK	N/A	OK	OK	N/A	N/A	N/A	N/A	N/A	N/A	OK	OK	so-so (C)	OK	down	OK	OK	OK	N/A
Halo 4	6/27/02	~15:40-19:50	C(2), S	OK	N/A	OK	OK	N/A	N/A	N/A	N/A	N/A	N/A	OK	OK	OK	so-so (7,12)	N/A	OK	OK	OK	N/A
CF 1	7/3/02	11:59-16:00	S, G, R	OK	N/A	OK	OK	OK	OK	OK	OK	OK	N/A	N/A	OK	OK	OK	N/A	OK	OK	OK	down
CF 2	7/3/02	17:50-20:53	R	OK	N/A	OK	so-so	down	OK	OK	OK	OK	N/A	N/A	OK	OK	OK	N/A	OK	OK	OK	down
CF 3	7/6/02	12:34-15:47	R, S	OK	N/A	OK	so-so	OK	OK	OK	OK	OK	OK	N/A	OK	so-so (C)	so-so (3)	N/A	OK	OK	OK	N/A
CF 4	7/7/02	12:23-15:37	S, C(3)	OK	N/A	OK	OK	OK	OK	OK	OK	OK	N/A	N/A	OK	OK	so-so (3)	N/A	OK	OK	OK	OK
CF 5	7/7/02	17:23-22:07	R	OK	N/A	OK	N/A	OK	OK	OK	OK	OK	N/A	N/A	OK	OK	so-so (3)	N/A	down	OK	OK	OK
CF 6	7/10/02	14:04-17:45	C(3), S	OK	down	OK	OK	OK	OK	OK	OK	OK	N/A	N/A	OK	OK	so-so (7)	OK	OK	OK	OK	OK
CF 7	7/11/02	15:25-20:09	G, R, T	down	down	OK	OK	OK	OK	OK	OK	OK	OK	N/A	OK	OK	OK	OK	OK	OK	OK	N/A
CF 8	7/13/02	17:25-22:21	C(8), G, R	down	so-so	OK	OK	down	OK	OK	OK	OK	OK	N/A	OK	OK	OK	OK	OK	OK	OK	N/A
CF 9	7/16/02	17:52-20:08	S	down	so-so	OK	OK	OK	OK	OK	OK	OK	OK	N/A	OK	OK	OK	OK	OK	OK	OK	so-so
CF 10	7/18/02	14:24-16:54	C(3)	OK	OK	OK	OK	OK	OK	OK	OK	OK	N/A	N/A	OK	OK	so-so (3)	OK	OK	OK	OK	so-so
CF 11	7/18/02	18:00-20:59	R	OK	so-so	OK	OK	OK	OK	OK	OK	OK	N/A	N/A	OK	OK	so-so (3)	OK	OK	OK	OK	down
CF 12	7/19/02	14:58-17:48	C(2), S	OK	OK	OK	N/A	OK	OK	OK	OK	OK	OK	N/A	OK	OK	OK	OK	OK	OK	OK	N/A
CF 13	7/19/02	19:01-23:07	R	OK	OK	OK	OK	OK	OK	OK	OK	OK	N/A	N/A	OK	OK	OK	OK	OK	OK	OK	N/A
CF 14	7/21/02	17:13-21:34	S, R	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK	N/A	OK	OK	so-so (3)	OK	OK	OK	OK	so-so
CF 15	7/23/02	19:29-23:53	R	so-so	so-so	so-so	OK	OK	OK	OK	so-so	so-so	N/A	N/A	OK	so-so (C)	so-so (3)	OK	OK	OK	OK	so-so
CF 16	7/25/02	14:00-18:09	C(2), S	OK	OK	OK	OK	OK	OK	down	OK	OK	N/A	N/A	OK	OK	OK	OK	OK	OK	OK	N/A
CF 17	7/26/02	15:56-19:59	S(2), C(1)	so-so	so-so	OK	OK	OK	OK	OK	OK	OK	OK	N/A	OK	OK	OK	OK	OK	OK	OK	OK
CF 18	7/28/02	18:31-22:34	R	OK	OK	OK	so-so	OK	OK	OK	OK	OK	N/A	N/A	OK	OK	OK	OK	OK	OK	OK	OK
CF 19	7/29/02	13:28-14:55	S	OK	OK	OK	OK	so-so	OK	N/A	OK	OK	OK	N/A	OK	OK	OK	OK	OK	OK	OK	OK
CF 20	7/29/02	17:00-21:10	R, I	OK	OK	so-so	OK	OK	OK	OK	OK	OK	N/A	N/A	OK	OK	OK	OK	OK	OK	OK	OK

Legend:

Flight types / maneuvers:

C = Cloud profiling of small/moderate cumulus

G = Coordination with ground RADAR site

I = Inflow run near large cumulus

R = Radiation runs under anvils

S = Spiral up/down for vertical profile (clear air)

T = Terra satellite overpass

Instrument performance:

OK Instrument was on and functioning

so-so Instrument suffered loss of data quantity/quality (instrument in parentheses is the non-functioning one)

down Instrument was not functioning

N/A Instrument was not on board or not turned on during flight

Instrument description:

CCN - CIT Cloud condensation nuclei counter (Caltech)

CCN - SIO Cloud condensation nuclei counter (Scripps)

Met

Nav

Meteorology probes (T, RH, P)

Navigational data system (C = C-migets, N = NovaTel, T = TansVector)

DMA	Differential mobility analyzer	CPC	Condensation particle counter (3 = UFCPC, 7 = 3010-24C, 12 = 3010-17C)
AMS	Aerosol mass spectrometer	APS	Aerodynamic particle sizer
CO	Carbon monoxide instrument	CAPS	Cloud, aerosol, and precipitation spectrometer
NOAA H ₂ O	NOAA water vapor instrument	FSSP	Forward scattering spectrometer probe
MACS	Multi-sample aerosol collection system	PCASP	Passive cavity aerosol spectrometer probe
SSFR	Solar spectral flux radiometer (part of Pilewskie package)	MOUDI	Micro-orifice uniform deposit impactor
BSR	Broadband solar radiometer		
LEISA	Linear etalon imaging spectral array		
ARI H ₂ O	Aerodyne water vapor instrument		